Effect of Size Press Coating of Cationic Starch/ Nanofibrillated Cellulose on Physical and Mechanical Properties of Recycled Papersheets

Ayhan Tozluoğlu [a] and Hakan Fidan [b,*]

This study aimed to evaluate the effect of size press coating on two types of recycled papers using different types of nanofibrillated cellulose (NFC) and cationic starch (CS) on physical and mechanical properties. Wheat straw was used as a cellulose source, and NFC was obtained through oxidative and enzymatic pretreatments. Recycled fluting and coreboard papers were coated with cationic starch/NFC blends at various concentrations of NFC (0.5%, 1%, 2%, 3%, and 4%) using a three-time repetitive size press application, followed by one-time drying section, and compared to uncoated papers. The application of a coating suspension containing 4% periodate-oxidized NFC on both paper surfaces resulted in a significant improvement in the tensile index, burst index, and internal bond strength of the papersheets, with increases of up to 60.6%, 96.3%, and 119.9%, respectively. Furthermore, significant decreases in air permeability were also observed with reductions of 75% and 81.6% for coreboard and fluting papers, respectively. Compared to other pretreatment methods, periodate oxidation resulted in higher viscosity values in NFC samples. Therefore, the application of periodate-oxidized NFC with a size press resulted in a significant improvement in the mechanical and barrier properties of papers made from recycled pulps.

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Keywords: Nanofibrillated cellulose; Recycled pulp; Papermaking; Coating; Size Press

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INTRODUCTION

In the papermaking industry, secondary fiber usage is increasing rapidly due to environmental consciousness and economical reasons (Sanchez-Salvador *et al.* 2020). However, the evaluation of recycled fibers in papermaking creates other challenges. The quality of the fibers gradually decreases with increased number of recycling cycles of paper because of mechanical and chemical treatments that occur during paper fabrication, and irreversible changes occur in the fiber structure, which is called hornification (Hubbe *et al.* 2007). Therefore, the use of recycled fibers is negatively affecting the physicochemical properties of fabricated final paper. Generally, beating of fibers and reinforcement additive usage are employed to improve mechanical and physical properties of recycled papers. Beating of pulp can potentially enhance the strength of final paper; nevertheless, recycled fibers are typically not subjected to beating due to the risk of excessive degradation, because recycled fibers have already undergone detrimental damage during the recycling process (Afra *et al.* 2013). Hence, to improve the strength properties of recycled pulp *via* increasing the fiber-fiber bonding ratio, various additives are commonly used, such as polyacrylamides (PAM), polyvinyl alcohols (PVA), cationic starch (CS), *etc.* (Hubbe 2006; Shen *et al.* 2021). Recently, the use of nanocellulosic materials (NC) as a reinforcing agent have gained increased attention from researchers and the industry because of their sustainability and environmentally friendly features (Hubbe *et al.* 2017; Poyraz *et al.* 2018; Li *et al.* 2021). Unique properties of NC, such as high aspect ratio, high specific surface area, tensile strength, low thermal expansion coefficient, and biological inertness, enable its usage in numerous fields as composites, electronics, scaffolds, drug delivery, food packaging, pulp and paper production, supercapacitors, *etc*. (Arslan *et al.* 2021; Barhoum *et al.* 2023; Mahardika *et al*. 2023). The term "nanocellulosic material" comprises various types, including nanocrystalline cellulose (NCC/CNC), nanofibrillated cellulose (NFC/CNF), and bacterial nanocellulose (BNC). Structural properties of isolated cellulose, such as morphology, thermostability, and crystallinity, primarily depend on the sources and processing conditions. For example, isolated microfibrils have long, thin, and flexible structure and contain both crystalline and amorphous domains, while crystalline parts left over after hydrolysis in strong acids are rod-like and rigid particles due to dissolution of amorphous regions. Amongst them, NFC holds a distinct advantage over CNC and BNC: it can be produced on a large industrial scale and offers a wide range of functional groups. Therefore, this makes NFC an excellent candidate for use as reinforcing agent in papermaking process. Experiments involving the utilization of NFCs in papermaking are not a recent development; they began several decades ago but were discontinued due to their lack of economic feasibility. Several drawbacks restricted the mass production and commercialization of NFC, such as high energy consumption (30,000 to 70,000 kWh/ton) and clogging during the homogenization. In the past few decades, many researchers have studied different pretreatment and isolation techniques to overcome high energy demand and clogging issues during the subsequent mechanical disintegration and enhance fiber delamination (Hubbe *et al.* 2017; Samyn *et al.* 2018; Candan *et al*. 2022).

After the development of pretreatment methods and the reduction in energy costs by a factor of over 100 times with the TEMPO-mediated treatment by Saito *et al.* (2006), the use of NFC in papermaking has been extensively investigated. The main focus of most research on NFCs in the papermaking industry has been using them as a bulk additive for virgin pulp and paper applications. The addition of NFC to the bulk suspension has a remarkable effect on the physical properties of paper. Because of its particle size and specific surface area, NFC fills and reduces the pores between fibers and increases the bonding ratio. As the concentration of NFC increases, the paper's density and opacity increase while its thickness, bulk, porosity, and air permeability decrease (Balea *et al*. 2019, 2020; Sanchez-Salvador *et al*. 2020). Nevertheless, its anionic charge restrains the retention of NFC on the paper web during paper web formation. Moreover, increasing NFC concentration and retention generally decreases drainage rate. For instance, González *et al*. (2012) reported that the bulk addition of 3% NFC to bleached eucalyptus pulp suspension increased °SR as 61.1%. Furthermore, 23.3% increase in °SR was reported by Tozluoğlu *et al*. (2021) after the bulk addition of 4% NFC to recycled pulp suspensions. The use of retention agents not only increases retention but also has positive effects on the mechanical properties by enhancing fiber-fiber bonding, thus proper selection and optimization of retention agents would be required.

In contrast, the application of NFC to already formed papersheets is another option to evaluate NFC in the papermaking process. The main advantage of this approach is that it completely eliminates the drainage and retention problems that occur during the papermaking process, as suspensions are applied after the paper fabrication (Espinosa *et* *al*. 2018). The use of cellulose nanofibrils in papermaking as coating agent is recent and has been subject of many investigations since the study by Syverud and Stenius (2009). The cited authors prepared MFC suspensions and coated papersheets using spray coating and reported an increase in tensile index and a dramatical decrease in air permeability for coated paper. Aulin *et al*. (2010) observed significant improvements in air permeability and oil resistance when carboxymethylated MFC was used to coat kraft and greaseproof paper using a rod coater. Similarly, Hassan *et al*. (2016) investigated the impact of NFC coating on the properties of bagasse paper sheets. They reported an increase in tensile strength and a decrease in porosity of the paper as a result of the NFC coating. The application of nanocellulose at the paper surface could be executed by several methods, such as rod coating, bar coating, dip coating, slot-die coating, roll-to-roll coating, spray coating, size press coating, yet the success and effectiveness of NFC coatings is mostly influenced from coating technique (Herrera *et al*. 2016; Kumar *et al*. 2018; Jin *et al*. 2021; Ozcan *et al*. 2021). It is possible to obtain different coat weights after certain solid content because coating techniques have different mechanisms. Lavoine *et al*. (2014) applied NFC as multilayers on papersheets using a size press and bar coating and examined the mechanical and barrier properties of the coated papersheets. The authors reported that a coat weight of 7 g/m^2 was achieved after applying five layers of NFC in bar coating, and this increased to 14 g/m^2 after ten layers. However, five layers NFC coating resulted in 3 g/m^2 coat weight in size press and increased up to 4 g/m^2 after ten layers. Both coating techniques resulted in a decrease of the air permeability and minor mechanical strength improvements. The authors also indicated that, because NFC suspensions consist of 98% water, wetting and drying cycles have a deteriorating effect on inter-fiber bonding of the paper. Similarly, Afra *et al*. (2016) investigated the deposition of a certain amount of NFC as 3 wt% single-layer and 1.5 wt% double-layer and reported that double-layer application of the same amount NFC demonstrated better results. The authors concluded that the application of a double-layer coating of 1.5 wt% NFC could potentially lead to a more uniform distribution of NFC on the paper surface. The first layer would fill the pores on the surface and, upon drying, form a relatively continuous film. This would create a smooth surface for the second layer to cover the surface without penetrating the paper bulk. NFC also has been used as an additive in various coating blends. For instance, in the study conducted by Hult *et al*. (2010), MFC was utilized along with shellac as a coating substrate. Their findings demonstrated significant enhancements in air permeability, but also indicated minor reductions in tensile strength. Lourenço *et al*. (2020) formulated a suspension of cationic starch and NFC with the aim of enhancing the printing quality of 80 g/m^2 office papers and observed improved surface properties of the coated papers. Amini *et al.* (2016) prepared NFC coating blends with silver nanoparticles (NFC/Ag) for food packaging and reported improved mechanical and barrier properties of coated papers in comparison with uncoated papers. Mirmehdi *et al*. (2018) prepared nanoclay/NFC coating substrate and applied it on a kraft paper *via* spray coating and formed hybrid composite layer on a paper surface and reported higher nanoclay content enhanced barrier properties but decreased tensile strength.

The present study aimed to assess the influence of different types of NFC as coating substrate on physical and mechanical properties of fluting and coreboard papersheets fabricated from various secondary fiber sources. For this purpose, wheat straw was used as primary fiber source to obtain NFCs. Before nanofibrillation, several pretreatments were employed: namely TEMPO- mediated (2,2,6,6-tetramethylpiperidine-1-oxyl) oxidation, PINO (phthalimide-N-oxyl) oxidation, sodium periodate ($NaIO_4$) oxidation, and enzymatic hydrolysis. Subsequently, coating suspensions were prepared *via* 4 wt% cationic starch and various proportions (0.5%, 1%, 2%, 3%, and 4%) of untreated and pretreated NFC and applied *via* a size press. The physical and mechanical properties of the base paper and starch/NFC-coated papersheets were evaluated, and the results were compared.

EXPERIMENTAL

Materials

Wheat straw (*Triticum aestivum* L.) was used as the raw material to produce nanofibrillated cellulose and harvested from Kütahya, Turkey. Fluting (110 g/m^2) and coreboard (160 g/m^2) papersheets were kindly supplied from Kahramanmaraş Paper Co. According to the supplier, fluting and coreboard papers were fabricated from recycled secondary fibers and surface-sized with 9 wt% gelatinized corn starch (13 cP) *via* size press. Three types of recycled papers were used for the production of fluting and coreboard papers (old newspaper and magazine papers (ONP, OMP) and mixed old corrugated cardboard pulp fibers (OCC), respectively). Pulping and bleaching chemicals (sodium hydroxide (NaOH), sodium borohydride ($NaBH_4$), sodium chlorite ($NaClO_2$), sodium acetate ($C_2H_3NaO_2$), acetic acid (CH_3COOH), and formic acid (CH_2O_2)) and pretreatment chemicals (TEMPO, PINO, and sodium periodate ($NaIO_4$)) were provided from Sigma-Aldrich (Taufkirchen, Germany). Endoglucanase and xylanase enzymes (Celluclast 1.5 L and Pulpzyme HC 2500) were kindly supplied from Novozymes (Bagsværd, Denmark). All chemicals were used without any further purification.

Pulping and bleaching

Soda-$NaBH_4$ cooking procedure was selected as pulping methodology, and pulping and bleaching sequences were conducted as reported by Tozluoglu *et al*. (2021). According to the procedure, cooking liquor was prepared at 14% active alkali by the addition of 0.5% of $NaBH_4$ o.d. (oven dried chips) with a liquor-to-wood ratio of 6:1 (L/kg). The cooking was conducted at 140 °C in a 10-L rotating digester (Uniterm Rotary Digester, Uniterm Laboratuvar Cihazları, Ankara, Turkey) for 80 min (40 min reaching maximum temperature, 40 min cooking) and the digester was stirred *via* a rotary motor during the cooking. Afterwards, the pulp was dispersed in a pulp disintegrator at 1200 rpm for 10 min and washed with tap water until it became black liquor-free, then screened using a laboratory-type screen according to TAPPI T275 sp-18 (2018) and yield was determined. Sodium chlorite bleaching was conducted to eliminate residual lignin. According to the procedure, the pulp was treated with a solution of 450 mL of $NaClO_2$ (15%) containing CH_3COONa, (3%), CH_3COO^-(7%), and CH_2O_2 (0.5%) at 5 wt% for 16 h. Later on, the pulp was washed with enough water to become neutral pH and stored for further applications. Bleached soda-$NaBH_4$ pulps were coded as AH.

Enzymatic hydrolysis

Bleached soda-$NaBH_4$ fibers were enzymatically hydrolyzed *via* commercially available endoglucanase (Celluclast 1.5 L) and xylanase (Pulpzyme HC 2500) enzymes, and enzymatic hydrolysis was performed as described by Sertkaya *et al*. (2023). Therefore, 50 g of bleached pulp (o.d.) was hydrolyzed in 2.5 L of phosphate buffer containing 11 mM KH_2PO_4 and 9 mM Na_2HPO_4 at 2% w/w solid loadings. The enzymatic hydrolysis was conducted at 50 °C for 2 h in an incubator (ES-20, Biosan Lab., Riga, Latvia) at neutral pH and xylanase and endoglucanase enzymes were loaded at 25-AXU/g and 2-EGU/g concentrations, respectively. Reaction mixtures were mixed every 30 min during the hydrolysis and, at the end of the 2 h, pulp was washed with distilled water and kept in a water bath at 80 °C for 30 min to terminate the enzymatic activity. Subsequently, hydrolyzed fibers were washed with distilled water repeatedly, and stored at 4 °C for further applications.

TEMPO-mediated (2,2,6,6-tetramethylpiperidine-1-oxyl) oxidation

TEMPO-mediated oxidation was conducted as reported by Poyraz *et al*. (2018). According to the procedure, 5 g of pulp (o.d.) was put in a beaker containing 500 mL of sodium phosphate (Na_3PO_4, 0.05 M) buffer prepared with 250 mg of NaBr and 25 mg of TEMPO at neutral pH. Then, 1.13 g NaCl (10 mM) and 0.5 mL NaClO (1.0 mM) were added to the suspension. The oxidation was completed at 60 °C for 72 h in a shaking incubator at 150 rpm. At the end of 72 h, to terminate the oxidation reactions, 100 mL of ethanol was added to the beaker and the mixture was filtered, washed with deionized water, and oxidized fibers were stored for further applications.

PINO (n-hydroxyphthalimide) oxidation

PINO oxidation was completed as described by Biliuta *et al.* (2011). According to the procedure, 16 g of pulp (o.d.) was added to a 1.4 L deionized water:acetonitrile (6:1) mixture. Subsequently, 3.2 mmol of anthraquinone (AQ) and 3.2 mmol phthalimide-N-oxyl (PINO) were added to the mixture, and the resulting suspension was kept in a room temperature (23 to 25 °C) for 5 days and continuously stirred. The reaction was terminated *via* addition of 100 mL ethanol to the suspension, and the mixture was filtered, washed with distilled water and acetone repeatedly, and oxidized fibers were stored for further applications.

Periodate (sodium m-periodate) oxidation

Periodate oxidation was applied as reported by Fidan *et al*. (2021). According to the procedure, 4 g of pulp (o.d.) was added to a beaker containing 266 mL of water solution prepared by 5.33 g of $NaIO_4$ and 15.6 g of NaCl, including the moisture of fibers. The reaction beaker was covered with aluminum foil to avoid light exposure and placed in a shaking incubator. The oxidation was conducted at room temperature for 96 h with continuous stirring at 105 rpm. At each specific reaction time (10, 16, 24, and 96 h), one-fourth of the mixture was exchanged with ethylene glycol to remove the residual periodate, thus halting the reaction. At the end of 96 h, the mixture was filtered, and oxidized fibers were washed with deionized water and stored for further applications.

NFC production

The nanofibrillation of the fibers (both untreated and pretreated soda-$NaBH_4$ pulps) was achieved *via* a high-pressure microfluidizer (M-110Y Microfluidizer, Microfluidics Corp., Westwood, MA, USA). For this purpose, 2 wt% aqueous suspensions of fibers were prepared and then passed through Z-shaped chambers by pressure following the next sequences: 1 time through a chamber with 200 μm diameter at 14000 psi, subsequently 5 times through a chamber with a 100 μm diameter at 24000 psi. The rheological properties of obtained NFC suspensions were studied in the authors' previous study (Tozluoğlu *et al*. 2021). The viscosity of NFC samples was determined with RST-CPS Rheometer (Brookfield Corp., Toronto, Canada) at room temperature. The measurements were made with the 37.5 mm diameter cone-plate and the 25 mm diameter parallel plate. The gap was fixed at 1 mm.

Prepared NFC samples were coded as their pretreatment and production methodology. Therefore, enzymatic hydrolysis, oxidation pretreatments, and nanofibrillation process were named as E, O, and n, respectively. Because the bleached soda-$NaBH_4$ pulp was coded as AH, as a result of nanofibrillation, untreated NFC was coded as AHn, enzymatically hydrolized samples were coded as AHEn1, AHEn4 (xylanase and endoglucanase, respectively), and oxidation pretreated samples were coded as AHOn1, AHOn2, and AHOn3 (TEMPO, PINO, and sodium periodate, respectively).

Surface sizing

Surface sizing was performed *via* self-designed size press machine as three-times repetitive application of coating suspension and one drying section. For this purpose, 100 mL of coating suspension was prepared containing different concentrations of NFC (0.5, 1, 2, 3, and 4 wt% to o.d. pulp) and 4 wt% gelatinized corn starch for each application. Subsequently, prepared suspensions were filled to size press and fabricated fluting, and coreboard papersheets were passed between rolls at 5 bar at a speed of 5 cm s^{-1} three times and dried with a contact drying system under 6 bar at 93 °C for 10 min. Each sample was conditioned at 50% relative humidity (RH) and 23 °C for at least 24 h according to TAPPI T402 sp-13 (2013) before testing. Physical (thickness, ISO 534 (1988); bulk, TAPPI T220 sp-01 (2001); and porosity, ISO 5636-3 (2013)), and mechanical (tensile index, TAPPI T494 om-01 (2006); burst index, TAPPI T403 om-15 (2015); internal bond, TAPPI T569 pm-00 (2000)), properties of the handsheets were determined according to the applicable standard methods.

All the data were statistically analyzed *via* analysis of variance (ANOVA) and Duncan’s mean separation tests.

RESULTS AND DISCUSSION

Physical Properties

It can be concluded from the results that physical properties were mostly affected in a negative manner, with the exception of air permeability. The thickness properties of fluting and coreboard papersheets coated with starch/NFC suspension using size press as a function of different NFC loadings are given in Fig. 1. In size press application, thickness is primarily affected by the gap between the rolls and applied pressure. However, viscosity also has an impact on these parameters at high solid loadings. If the pressure falls below a certain level, coating suspension may not spread evenly, thus leading to an uneven coating (Boissard 2017). Because the used base fluting and coreboard papers were already surface-sized with cationic starch, subsequent size press application increased their thickness to a certain extent, and as expected, the thickness values were even higher with increasing NFC loadings. The thickness values of base fluting and coreboard papers were 294 μm and 330 μm, respectively. The size press application not only forms a coating layer to the surface, but it also allows penetration of the coating suspension beneath the paper surface with pressure. This results in a lower thickness increase compared to other coating applications (such as bar coating, spray coating, *etc*.), and the observed thickness increase is generally referred to swelling of the paper as water exposure damages fiber-fiber bonds. A comprehensive study was carried out on this matter by Lavoine *et al*. (2014), who stated that 10 times application of 2% MFC suspension on papersheets *via* bar coating has no significant effect on thickness when compared to only water application. They also reported that there is no significant increase in thickness between 5 times and 10 times the size of press application of MFC suspension. The researchers concluded that because of the high water content of the suspension (98%) and the pressure applied during size press, the coating suspension was able to penetrate the substrate, resulting in an inability to achieve a uniform coat layer, even with 10 times applications of the size press.

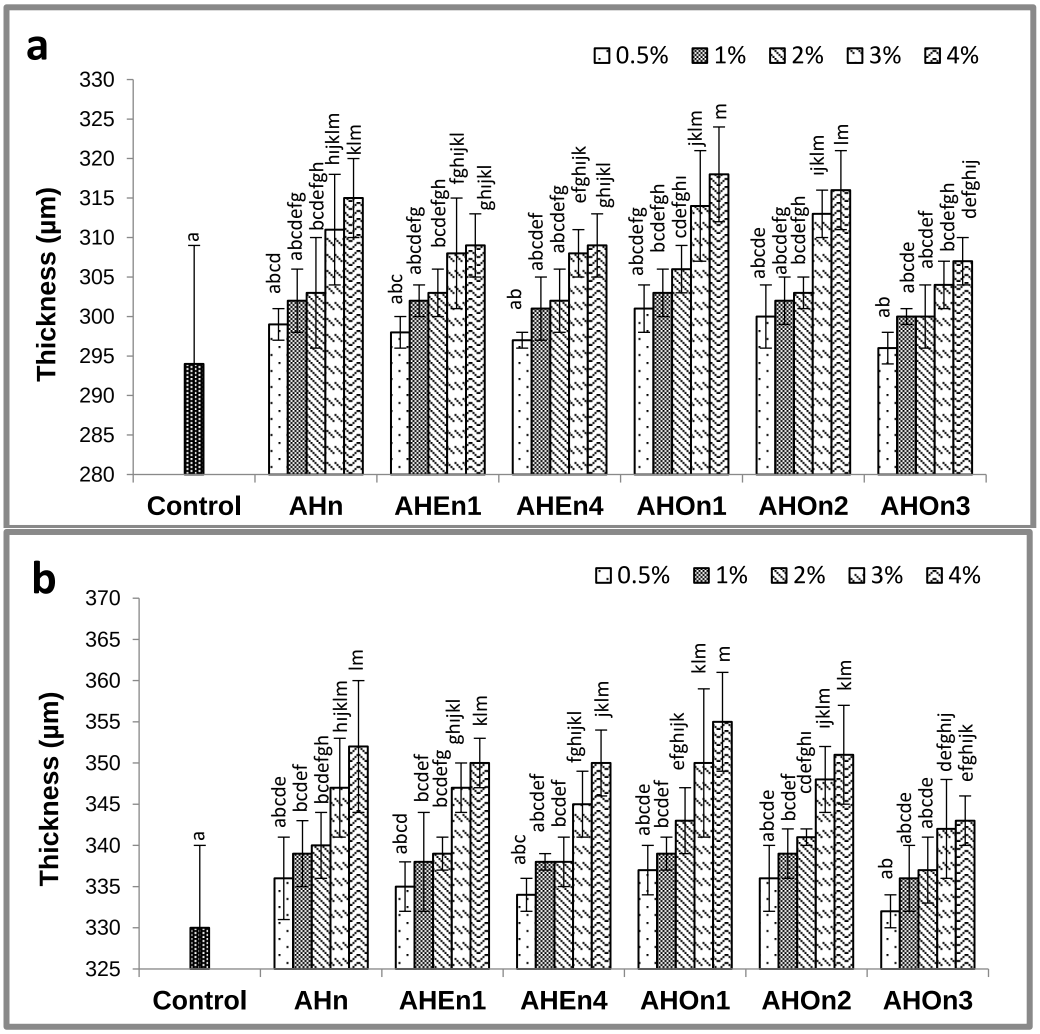

Fig. 1. The effect of starch/NFC suspensions on the thicknesses of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

As stated above, viscosity of the coating suspension has considerable effect on the achieving uniform coating layer. Studies have shown that NFC suspensions with higher viscosity exhibit reduced penetration into the base paper (Sharma *et al*. 2020). Given that increasing the concentration of NFC leads to higher viscosity, it is expected to result in a decrease in the permeation of NFC fibrils into the paper structure. Furthermore, the use of starch and other binders in NFC coating suspensions has positive effects of viscosity. Mazhari Mousavi *et al*. (2017) indicated that incorporating CMC as an additive to alter the rheological properties of NFC suspension, by decreasing the fiber-fiber interactions of

NFC fibrils, thus reduces flocculation tendency of NFC suspensions; therefore, it enables uniform spreading of NFC at moderate solids content. The rheological behaviours of the evaluated nanofibrillated celluloses in this work were given in the authors' previous study (Tozluoğlu *et al.* 2021). The periodate-oxidated samples exhibited lower viscosity than TEMPO and PINO oxidated samples, therefore the formulation was able to penetrate and uniformly spread on the substrate surface under high pressure. Therefore, AHOn3 samples resulted in a lower increase in thickness values of both fluting and coreboard paper even at 4% NFC loadings (13 to 25 µm, respectively). In contrast, Boissard (2017) reported a 3 µm increase in thickness of paper after one-time size press application of NFC, whereas Lavoine *et al.* (2014) reported 6 µm increase even 10 times size press application of NFC. Additionally, bar coating of 300 g/m^2 papersheets *via* 2% MFC suspension determined a 50 µm increase in thickness after 10 times successive coating and drying application. In the current coating procedure, starch is incorporated as a mixture component in the coating suspension and it was applied once to prevent water damage. As a result, higher increases were observed in thickness compared to the findings in the literature, possibly due to a higher deposition rate of starch and NFC on the substrate surface.

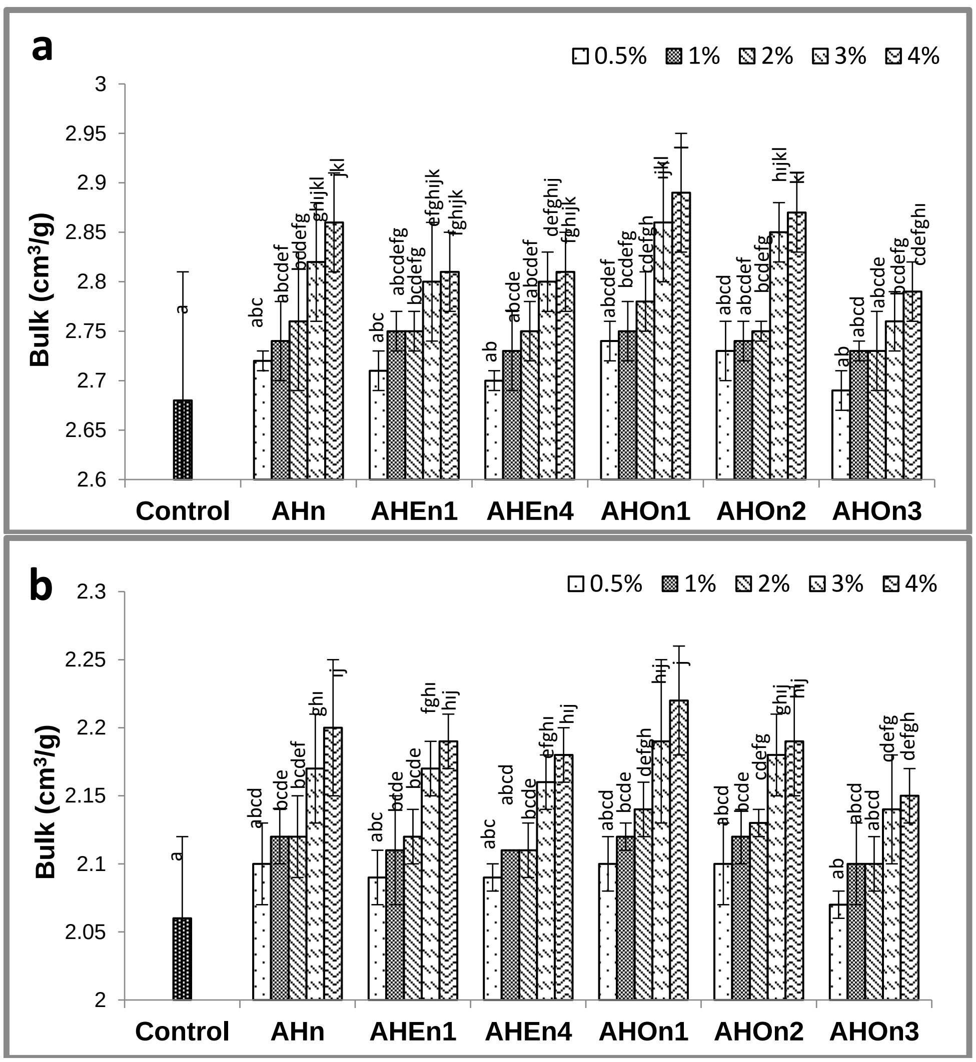

Fig. 2. The effect of starch/NFC suspensions on the bulk of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

The bulk values of the surface-sized paper sheets are given in Fig. 2. Because coating applications generally increase the thickness, unlike the bulk addition, as expected the surface sizing of starch/NFC suspensions increases the bulk values of paper (Balea *et al*. 2016). It is concluded that there is a direct correlation between the measured paper bulk and viscosity of the prepared starch/NFC suspensions. Additionally, the usage of recycled fibers contributes to this phenomenon as well.

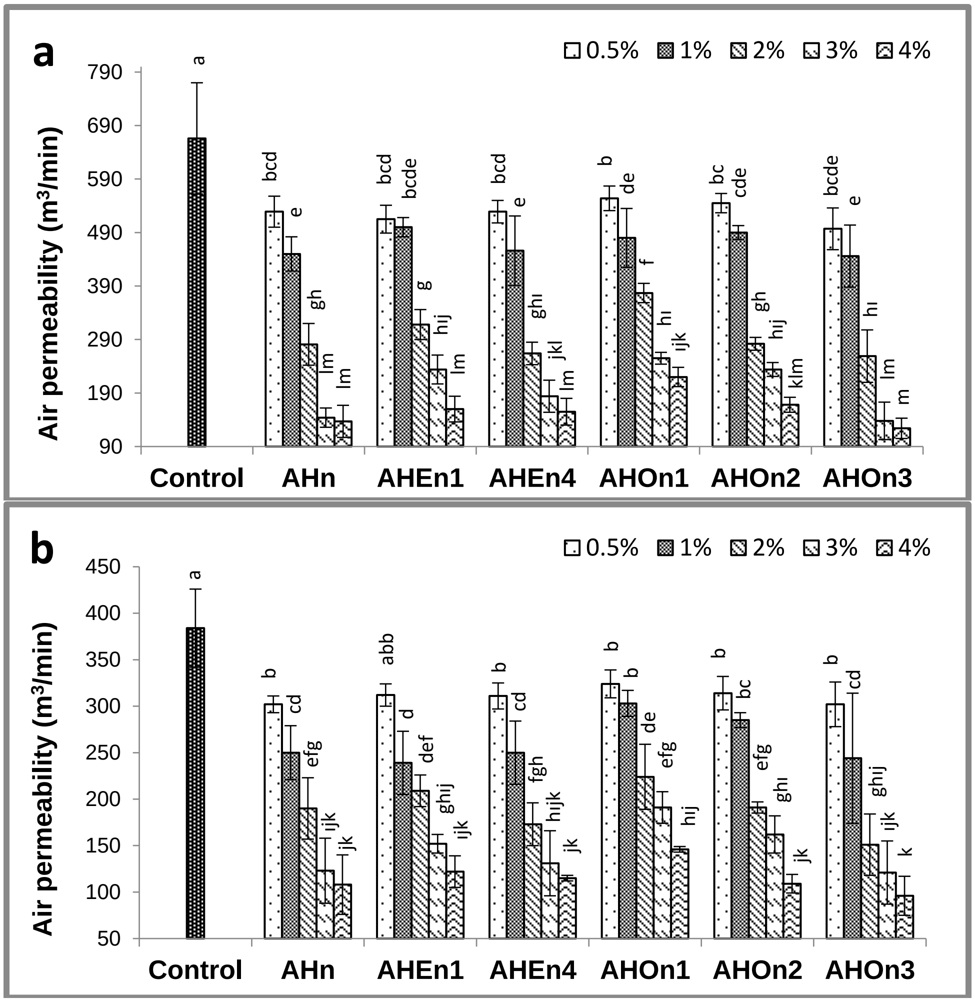

Fig. 3. The effect of starch/NFC suspensions on the air permeability of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

Due to the filler content of recycled fibers, the fabricated paper demonstrated a porous structure. Initial surface sizing application *via* starch was insufficient in filling the gaps in the paper. As a result of surface sizing applications, the highest increases were observed in AHOn1 (TEMPO-treated) samples at a 4% concentration for both paper types, with increases of 7.9% and 7.5% observed in fluting and coreboard, respectively ($p < 0.001$). Because AHOn3 (perioated-treated) samples showed lower viscosity, their suspensions penetrated beneath the surface, resulting in lower bulk increase for AHOn3-coated fluting and coreboard paper sheets (4.10% and 4.37%, respectively). These results are in line with previous studies reported in the literature. In a study conducted by Lavoine *et al*. (2014), CMF suspensions were applied to 300 g/m^2 *via* bar coating process. The results indicated that the bulk value of the cardboard increased with each additional coating, and when the coating was applied 10 times, the bulk value increased 19.4% compared to uncoated cardboard (from 1.65 cm^3/g to 1.97 cm^3/g). In another study, paper sheets made from different fiber blends (eucalyptus, sisal, and pine) were coated with 10% NFC suspensions. The results indicated that coating NFC onto paper was more effective in increasing bulk than adding the same amount of NFC to pulp slurries (Pego *et al*. 2020).

Air permeability of paper coated with starch/NFC suspensions using the size press as a function of coat weight is given in Fig. 3. As expected, the air permeability was significantly decreased as a result of surface sizing of various starch/NFC suspensions onto paper ($p < 0.001$). Air permeability of control paper was measured as 384 m^3/min for coreboard and 666 m^3/min for fluting paper and, after the size press applications, it was determined in the range of 324 to 96 m^3/min and 554 to 124 m^3/min, respectively. In both paper types, periodate-treated samples demonstrated lower air permeability values at 4% NFC concentrations, a decrease of 75% for coreboard and 81.6% for fluting papers. It is worth mentioning that the air permeability here is affected by both the level of porosity of the base paper and applied coating technique. There are two possible explanations of this phenomenon. One is that the starch/NFC suspensions form a proper layer on the paper substrate, the other is that lower viscosity allowed suspensions to penetrate beneath the surface and fill the gaps between the fibers, therefore improving the air resistance of papersheets. Even though recycled fiber sources have non-fiber materials such as fillers in their structures, base paper demonstrated low air permeability values compared to the previous studies because the paper substrates were already surface sized with starch. In a similar study, Lavoine *et al*. (2014) did not find any significant improvement in air permeability of commercially coated 300 g/m^2 paper after 10 times successive bar coating of NFC suspensions. However, Fidan *et al*. (2021) reported a 69.8% decrease in air permeability of 90 g/m^2 fluting papers after size press application of 4% starch/NFC suspension. In contrast to the size press, El-Samahy *et al*. (2017) reported that bar coating of the surface of 110 g/m^2 papersheet with NFC decreased the air permeability up to 7 times depending on the thickness of the coating. Moreover, Liu *et al.* (2014) reported 100 times decrease in air permeability of papersheets after bar coating process. In another study, cellulose nanocrystals were combined with carboxymethyl cellulose (CMC) and silver nanoparticles to form an antibacterial coating. The authors observed a 93.3% decrease air permeability after 7% concentration of CNC (He *et al.* 2021). However, significant decreases in air permeability (33.03%) were observed even in low solid loading (1 wt%) in the present study. Boissard (2017) suggests that the air permeability of substrates coated with a size press was lower than that coated with a bar coating, especially at low solid contents. This statement suggests that despite not forming a complete layer on the paper surface *via* size press application, the NFC coating still results in a lower overall porosity of the substrate by filling the gaps between fibers.

Differences in the mean thickness, bulk and air permeability values for all starch/NFC-treated samples were statistically significant for both fluting and coreboard pulp papers ($p < 0.001$).

Mechanical Properties

A remarkable increase in the mechanical properties of starch/NFC-coated samples was observed, especially at higher solid loading compared to the uncoated samples. It can be concluded that the NFC loadings and their viscosity degrees were directly correlated with overall mechanical properties. Surface sizing of paper with various proportions of starch/NFC suspensions resulted in a significant increase in tensile indices compared to the starch-coated samples, and with the increasing NFC concentrations, tensile indices increased gradually. Starch-sized paper demonstrated 49.3 Nm/g for fluting and 48 Nm/g for coreboard samples. Applying starch/NFC at different proportions to paper resulted in an increase in the range of tensile index values from 53.3 to 75.6 Nm/g for fluting paper and 54.8 to 77.1 Nm/g for coreboard paper. In both fluting and coreboard papersheets, the highest increase in tensile indices was observed by applying 4 wt% of AHOn3 samples as 53.4% and 60.6%, respectively ($p < 0.001$).

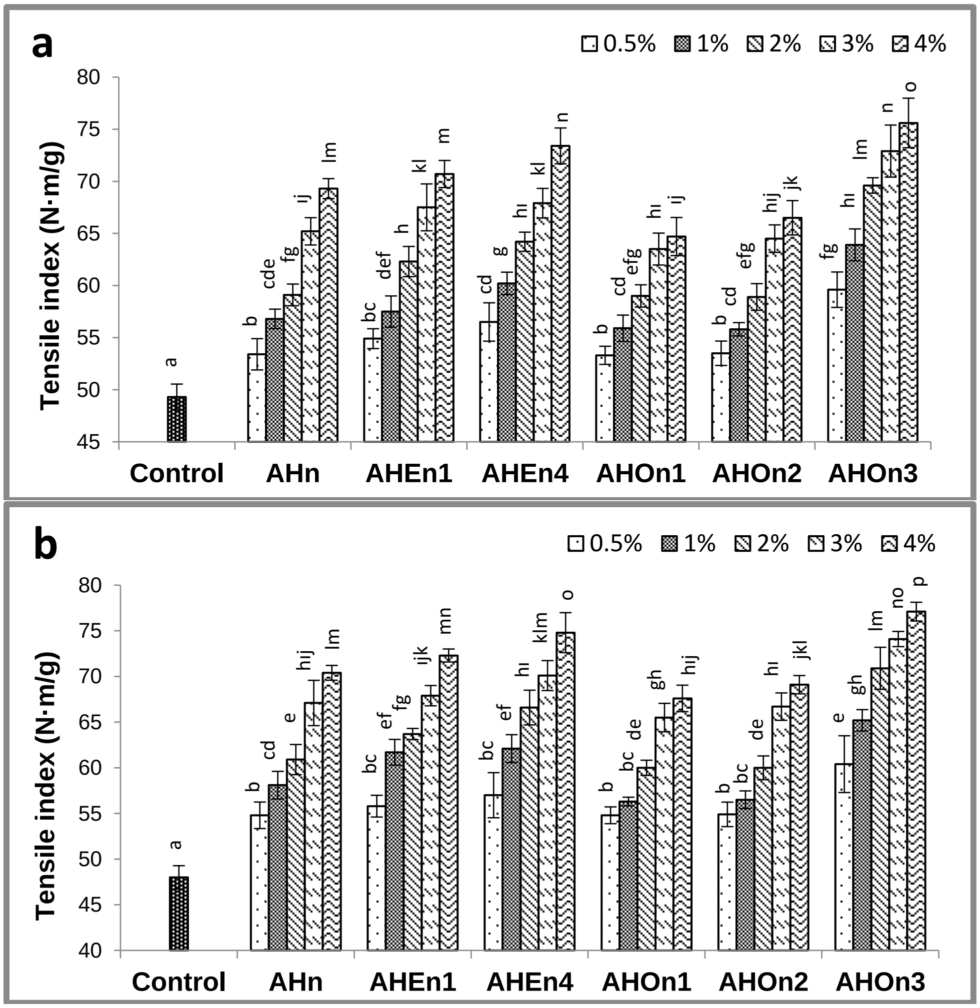

Fig. 4. The effect of starch/NFC suspensions on the tensile indices of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

Due to its lower viscosity, periodate-treated samples can distribute uniformly in a starch medium, and with the pressure, the suspension can penetrate under the surface, which leads to an increase in fiber-fiber interactions and consequently higher increase in tensile indices. This phenomenon was also confirmed by Fidan *et al*. (2021). According to their study, applying 4% of periodate-treated NFC to 90 g/m^2 fluting papersheets increased the tensile index 130% compared to the base paper and 52.1% compared to the only starch-coated paper. However, the impact of NFC coating on tensile strength of paper is a controversial topic in the literature (Brodin *et al.* 2014). Some studies suggest that the addition of NFC may lead to negligible improvements or even slight decreases in the tensile index (Syverud and Stenius 2009). Mazhari Mousavi *et al.* (2017) prepared coating suspension with NFC and CMC (carboxymethyl cellulose) and bar-coated 200 g/m^2 paper as double layers. The authors reported maximum 10% and 20% increases in tensile index at 4% NFC loadings in machine direction and cross direction, respectively. They suggested that this low improvement could be explained by the surface properties of the paperboard. The pores and holes on the surface may lead to discontinuity and contribute to disorder in NFC layers made on the surface of paperboard. Additionally, Kumar *et al*. (2018) reported a 10% increase in tensile index of 170 g/m^2 paperboard at coat weight higher than 10 g/m^2. In another study, NFC was used as coating suspension to carry triclosan to use as an antibacterial coating and at 30% solid loadings, and only 18% increase in tensile index is observed Liu *et al.* (2015). In contrast, Mirmehdi *et al*. (2018b) reported 41% increase in tensile index of 75 g/m^2 paper after applying 1.4% NFC *via* spray coating. It should be noted that most of the studies were conducted on virgin pulp-based paper. Relatively higher results in the present study could be explained by the use of recycled fiber to fabricate paper. Furthermore, initial starch sizing influenced the results as well.

The burst strength is directly related to the inter-fiber bond strength as well as individual fiber strength. In addition, evaluated fiber source also affects the burst strength whether it is virgin fiber or secondary fiber (Hubbe 2006). Generally, recycled fibers show lower mechanical properties when compared to virgin pulp due to the filler content and the hornification of fibers, and it is expected that the application of surface sizing would increase burst strength by incorporating fibers *via* cationic starch (Sanchez-Salvador *et al*. 2020). The burst index values of the papers surface sizing with starch alone were determined as 2.15 kPam^2/g and 2.43 kPam^2/g for fluting and coreboard papers, respectively. As shown in Fig. 5, surface sizing of starch/NFC suspension to fluting and coreboard substrates gradually increased burst indices in the range of 2.74 to 3.74 kPam^2/g for fluting and 3.36 to 4.77 kPam^2/g for coreboard papers. For coreboard samples, size press application of 4% NFC nearly doubled the burst index values. Because inter-fiber bond strength significantly influenced the burst strength, it can be concluded that coating suspension penetrated under the paper surface and contributed to increase fiber interactions of paper web.

Cuong *et al*. (2021) reported 5.8% and 8.8% increases in burst strength of 125 g/m^2 paper with the bulk addition of 2% and 5% rice straw NFC, respectively. In the same manner, Tajik *et al*. (2018) fabricated 60 g/m^2 bagasse papers by the bulk addition of 2 wt% NFC and reported only 18% increase in the burst index. It is often stated that superficial applications of NFC results better on burst strength, in contrast to bulk additions (Adel *et al.* 2016; Balea *et al*. 2016; Fathi and Kasmani 2019). The main reason for this phenomenon is the ability to adjust the NFC concentrations precisely in surface applications and to avoid retention issues in post-fabricate applications. Kumar *et al*. (2016) treated 178 g/m^2 kraft paper *via* roll-to-roll coating with 2% NFC/CMC suspensions and reported an increase in burst strength up to 50%. Similarly, in a study conducted by Fidan *et al*. (2021) remarkable increases are observed in burst strength after size press application of starch/NFC suspension to 90 g/m^2 fluting papers. Authors reported 124% increase in burst strength *via* 1 wt% starch/NFC suspension and up to 173% at 4% NFC concentrations. In the present study, similar results were observed.

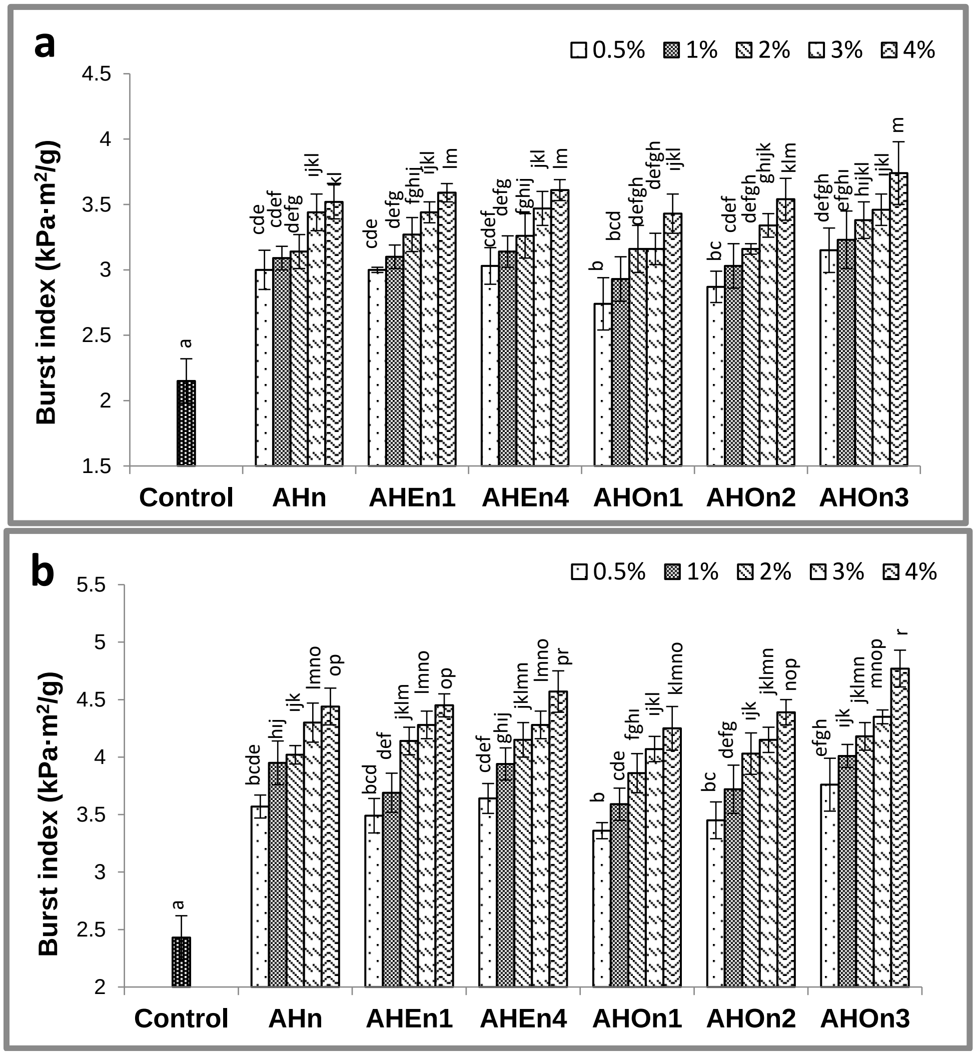

Fig. 5. The effect of starch/NFC suspensions on the burst indices of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

AHOn3 samples (periodate-treated samples) exhibited increases of 46.5% and 54.7% at 1% NFC loading for fluting and coreboard papersheet, respectively. Furthermore, these increases reached up to 73% and 96.2% when NFC concentrations increased to 4 wt%. In contrast, Lavoine *et al*. (2014) indicated that there were no significant changes in burst strength, and even slight decreases are observed after repetitive bar coating and size press applications. It was stated that the decreases could be attributed to the wetting and drying cycles, which have a negative impact on the inter-fiber bonding. Considering that NFC suspension primarily consists of water (98%), and coating applications involve repeated cycles, exposure to water could lead to the rupture of H-bonds within the paper structure, thus negatively affecting overall mechanical strength. As mentioned above, burst strength was directly related to inter-fiber bonding ratio and bond strength. Therefore, one-time wetting and drying cycle in the present study has not affected the bond strength, and the starch usage along with the NFC as suspension gradually increases burst strength by enhancing fiber-fiber bonds and forming entangled network structure.

The internal bond strength of paper has significant importance, as it directly correlates with the bonding level between fibers and their overall strength. Besides, poor internal bonding can result in separation and delamination issues during processing of the paper. There are several parameters that affect internal bonds including base paper strength. Papers fabricated from recycled fibers exhibit lower mechanical properties when compared to the virgin pulp-based, leading the coating processes to be more influential on mechanical properties of recycled papers (Espinosa *et al*. 2019). Therefore, as expected, surface sizing application of starch/NFC suspensions significantly increased the mechanical properties of the fluting and coreboard paper, and the incremental effect was further increased in higher NFC ratios.

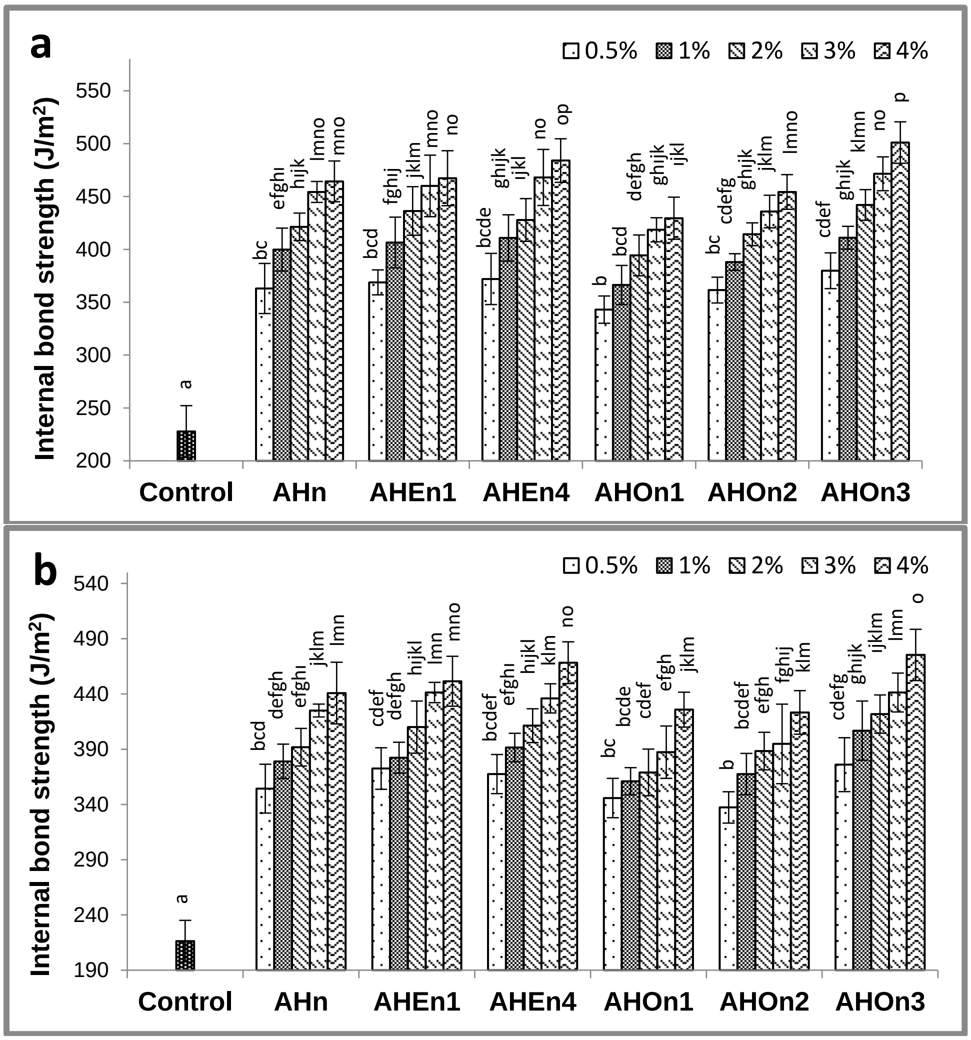

Fig. 6. The effect of starch/NFC suspensions on the internal bond strength of (a) fluting and (b) coreboard papers. Factors followed by the same letter were not significantly different (Duncan test at $p < 0.05$).

Internal bond values of control paper were observed as 227.8 J/m^2 for fluting and 216 J/m^2 for coreboard samples. After the surface sizing application of different proportions of starch/NFC suspensions, remarkable increases were observed (up to 119.9%) and the highest increases were obtained in AHOn3 samples for both samples. This finding is mentioned in previous studies, stating that periodate treatment reduces NFC viscosity, allowing NFC to penetrate not only the surface but also the subsurface of the fibers (Poyraz *et al*. 2018). Additionally, Lavoine *et al*. (2014) noted that size press application leads suspension to penetrate under the surface and enhances fiber-to-fiber interactions by increasing H-bonds. When considering these situations, an increase in internal bond strength after size press application is inevitable.

In addition, apart from the viscosity of suspensions, the texture of the base papers also plays a big role in this phenomenon. Papers fabricated from recycled fibers demonstrate porous structure, thus facilitating the penetration of coating suspensions beneath the surface. Therefore, even TEMPO-treated samples (AHOn1) exhibited higher viscosity values, at 1% NFC loadings, 60.8% and 66.9% increase in internal bond strength is observed for fluting and coreboard samples, respectively. Similarly, Tarrés *et al*. (2016) reported 119.0% increase in internal bond strength of 75 g/m^2 paper coated with 0.45 wt% NFC and 2.5 wt% cationic starch. Furthermore, Fidan *et al*. (2021) observed 110.9% increase in internal bond strength after surface sizing of 90 g/m^2 fluting papers *via* starch/NFC suspensions. In contrast, Beneventi *et al.* (2014) reported a slight decrease in internal bond strength of 35 g/m^2 kraft paper samples after application of 6 g/m^2 MFC *via* spray coating. It was stated that the decrease could be explained by the low grammage and porous structure of evaluated samples. Contrastingly, Ottesen *et al*. (2017) applied 0.85 wt% TEMPO-NFC/5 wt% CMC coating suspension on 255 g/m^2 recycled paperboards using slot-die method and reported 27.8% increase in internal bond strength. It was indicated that high grammage of the paper and low solid loadings may diminish the incremental coating effect and resulted in a relatively low increase in internal bond strength. Compared to the findings in the literature, this gradual increase can be attributed to the high solid loadings of the coating suspensions. Furthermore, the size press allowed the coating suspension to penetrate the substrate due to the porous structure of recycled paper, thus resulting in a significant increase in the internal bonding strength.

Differences in the mean tensile index, burst index, and internal bond strength values for all starch/NFC-treated samples were statistically significant for both fluting and coreboard pulp papers ($p < 0.001$).

CONCLUSIONS

1. Superficial application of starch/nanofibrillated cellulose (NFC) suspensions improved overall mechanical properties of recycled fluting and coreboard paper.
2. Using cationic starch along with the NFC as viscosity modifier led to better distribution as well as better retention of NFC suspensions onto paper surfaces.
3. Size press application of starch/NFC suspensions increased the tensile indices of fluting and coreboard paper up to 53.4% and 60.6%, burst indices up to 74.0% and 96.3% and internal bond strength up to 119.9% and 119.7%, respectively.
4. Air permeability of surface-sized coreboard and fluting papersheets decreased as 75.0% and 81.4% with the surface application of 4 wt% AHOn3 samples due to periodate oxidation yielded in lower viscosity amongst other pretreatment methods.
5. It can be concluded that the post-fabrication applications of NFC to paper gives better results than bulk addition due to eliminating the drainage and retention problems that occur during the papermaking process.

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